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ULTRASONIC TEMPERATURE MEASURING DEVICE

by

L. C. Lynnworth and E. H. Carnevale

prepared for

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

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PARAMETRICS, Inc.

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ABSTRACT

Thirty wire samples of seven different refractory elements and alloys were irradiated in the Babcock & Wilcox test reactor. Subsequently, the effect of radiation on the sound velocity-temperature relationship will be determined. Materials have been reviewed with respect to sensor and sheath applications. It appears that pure Re is the preferred sensor, and tungsten, the preferred sheath, for use in graphite up to at least 4500°R. Other combinations are required for possible use to 5000°R and above, depending on time at temperature, etc. The automatic ultrasonic thermometer instrument has been tested with Re, W and Mo wires heated up to about 4200°R, with satisfactory results.

Author

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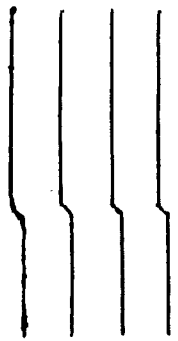
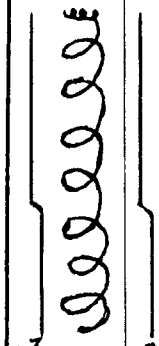
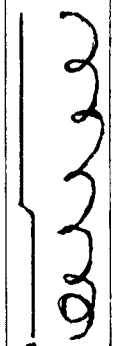
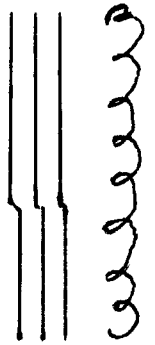
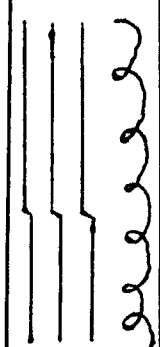
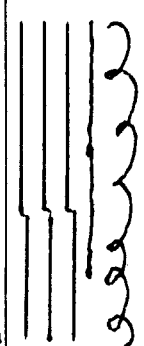
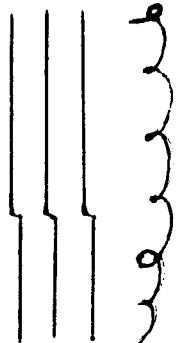
I. RADIATION TESTS

The thirty encapsulated samples of seven different refractory elements and alloys (Table I) have now been irradiated as required. The capsule was opened in a hot cell approximately one month after pulling. Measurements of radiation levels will determine how soon individual wires can be shipped from Babcock & Wilcox to us, for further high temperature tests. Had the wires been totally free of contaminants, Cb and Mo could have been shipped at once, and Re, one month later. Ta, W, and W-Re alloys are being stored at Babcock & Wilcox. Initial qualitative radiation measurements at Babcock & Wilcox, however, showed traces of Co and Ta in Cb and Mo, and other impurities in Re. These results, not unanticipated, were discussed at Parametrics with J. Wett, Jr. of Babcock & Wilcox. It was concluded that further measurements in Re are required, to identify and measure the radioactive isotopes. These measurements will be performed by Babcock & Wilcox or Parametrics. Subsequently, high temperature ultrasonic tests will be conducted on short specimens of irradiated wires (~2 inches) instead of testing complete lengths at once (see Table I). These ultrasonic tests will be conducted by Parametrics, either at Babcock & Wilcox or in our own laboratory. Results will be compared with our own pre-irradiation tests, as well as with other independent data[1, 2] (see, for example, Figure 1).

Table I. Summary of Wires Inserted into Babcock & Wilcox Reactor

MATERIAL	TUNGSTEN	RHENIUM			TANTALUM	MOLYBDENUM		COLOMBIUM	W-5%Re	W-26%Re
Diameter, inches	0.04	0.02	0.03	0.04	.04	0.02	0.04	0.02	0.02	0.02
Wire Shape										
Wire Length, inches (approx.)	5	$4\frac{7}{8}$ 30	$4\frac{7}{8}$ 24	$4\frac{7}{8}$	4-$\frac{5}{8}$ 120	4-$\frac{1}{2}$ 120	4-$\frac{1}{2}$ 120	5 120	$4\frac{3}{4}$ 24	$4\frac{5}{8}$ 24
Total number of wires of each material	4	5			4	9		4	2	2

II. REVIEW OF SENSOR MATERIALS

In this section, we briefly review sensor materials such as graphites, carbides, polycrystalline metals and single crystal metals.

A. Graphites

To minimize or avoid reactions with the graphite core, graphite is a natural first choice as a candidate sensor material. However, Faris, Green and Smith's early work in AUF and SA-25 graphites[3] (Figure 2), Johnson and Dull's more recent work[4] in an unspecified "highly anisotropic graphite" (Figure 3), and Armstrong and Brown's recent work[1] in AUC, G and H4LM graphites (Figure 4), indicate that sound velocity varies considerably from sample to sample, and that the velocity is often insensitive to temperatures over wide ranges, and also two different temperatures yield the same velocity (Figures 3, 4). Bell, Hub and Smith[5] tested CS and EY9 graphites up to about 1100°R and observed hysteresis effects which they attributed to water absorption (Figures 5, 6). Additional hysteresis effects are to be expected near 5000°R due to creep and annealing processes. Available Young's modulus data[6] for pyrolytic graphite (PG) indicates that sound velocity is not very sensitive to temperature in the range 3000 to 4000°R (Figure 7).

B. Carbides

After graphite, the second choise material likely to withstand the graphite environment would appear to be one in which the carbide reaction has gone to completion, i.e., a carbide.

Unfortunately, the task of fabricating a carbide which will remain stable at 5000°R is difficult. Moreover, reliable modulus data is meager and scatter in the data even at room temperature is substantial (Young's modulus in WC = 75 to 103 million psi).[7] Although the carbides generally have rather high melting points, Table II shows[7] that the maximum use temperature in the present carbon/hydrogen environment is generally below 5000°R.

Carbides tend to be unstable near 5000°R. Depending on temperature, pressure and hydrogen flow conditions, the net reaction may be either decarburization or formation of a higher carbide. The effect

Table II. Comparison of Some Carbides*

Carbide	M. Pt., °R	Reactivity/Mechanical Properties	Max. Use Temp. in Graphite/Hydrogen Environment, °R
MoC	5100-5250	Decomposes to Mo_2C at 2100°R	2100
Mo_2C	4800-5400	Decarburizes rapidly in H_2 above 3200°R	3200
TaC	7200-7300	Attacked by H_2 above 3200°R	3200
WC	5200-5700	Decarburizes above 3200°R in H_2	3200
W_2C	5400-5700	Decarburizes in H_2, hydrocarbons above 5000°R	5000 (?)

*Data and remarks extracted from Shaffer (ref. 7)

of composition on sound velocity is illustrated in Table III for W, W_2C, WC and C (diamond), calculated at room temperature from Köster and Rauscher's data.[8] Velocities in MoC and Mo_2C were also calculated from Shaffer's data.[7] Uncertainties in composition as severe as these could lead to temperature errors of thousands of $^{\circ}R$. Even in a given carbide there are uncertainties. For example, in W_2C, Schwarzkopf and Kieffer[9] point out that the transformation from β - W_2C into α-W_2C occurs near 4800$^{\circ}R$. The lattice structure of the modification stable at high temperatures had not been fully established at the time of that writing (1953).

Another problem relevant to a carbide sensor is the difficulty of handling pure carbides in lengths greater than about 5 diameters. Pure carbides are also characterized by poor thermal shock resistance.*

In the case of near stoichiometric UC, sound velocity is not sensitive to temperature over the range 500 to 2000$^{\circ}R$, and attenuation increases above 2000$^{\circ}R$, in a nonreproducible manner.†

In summary, the presently available carbides, despite attractively high melting points, are generally unsuited for reliable use from 1000 to 5000$^{\circ}R$, as a thin wire temperature sensor.

C. Polycrystalline Metals

Polycrystalline refractory metals are readily available in wire form in relatively unlimited lengths, except for Re, which is available in lengths of ~5 feet in diameters of ~0.040 in. The Dragon Project concluded that Re was the best sensor material, and our results to date confirm this. However, on tests conducted with 0.030 inch diameter bare Re sensor wire in intimate contact with graphite, attenuation unexpectedly increased near 3500$^{\circ}R$ with some recovery of signal observed on cooling to room temperature. Embrittlement also occurred, as expected.[2] Similar tests on a second specimen again showed a marked increase in attenuation at 3500-3800$^{\circ}R$, with recovery of signal on cooling.

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*R. DeCesaris, Chief Metallurgist, Kennametal, private communication (July 18, 1966).

†G. Ervin, Jr., Atomics International, private communication (August 17, 1966). See Atomics International Quarterly Prog. Rpt. 7551-4539, p. 2 (April 6, 1965).

Table III. Effect of Composition on Extensional Wave Sound Velocity in Carbides of Tungsten and Molybdenum at Room Temperature*

Material	Young's Modulus, psi	Density, g/cc	Extensional Wave Sound Velocity in./μsec
W	59×10^6	19.3	0.181
W_2C	62×10^6	17.3	0.195
WC	104×10^6	15.8	0.265
C(diamond)	115×10^6	3.5	0.592
Mo	46×10^6	10.2	0.214
Mo_2C	32.5×10^6	8.9	0.193
MoC	29×10^6	7.6 (pycn. density = 8.5-8.8 g/cc)	0.197

*Velocity calculations based on data in Shaffer (ref. 7) and Köster and Rauscher (ref. 8).

Room temperature velocity was virtually unaffected by carbon diffusion, but the velocity at elevated temperature was affected by the diffusion process. The Re-C phase diagram[2] shows 4.5 at. % carbon in the range 3500-4000°R, increasing to 11.7 at. % carbon at the 4964°R eutectic. To avoid the effects of carbon diffusion into rhenium, adequate sheathing is required.

D. Single Crystal Metals

Single crystal metals offer several potential advantages over polycrystalline metals: (1) No grain reorientations during thermal cycling; (2) More linear velocity/temperature curve; (3) No grain boundaries for carbon attack, slip, etc. Disadvantages are: (1) Length limited; (2) Diameter limited; (3) Machining O.D. is difficult; (4) Cost is high, ~$100 to $200 per inch (however, use of short single crystal sensor butt welded to polycrystalline lead-in would be reasonable in cost).

E. Sensor Materials for Use Beyond 5000°R

Although temperatures above 5000°R are beyond the scope of the present contract, recent interest in such temperatures warrants brief consideration here. PG exhibits a sound velocity which is particularly sensitive to temperature in the range 4500 to at least 6000°R. Other forms of graphite might also be suitable for that range.

Figure 8 compares the reciprocal velocity in some of the above mentioned materials to the extent that data are available. (The upper and lower W curves represent poly- and single-crystal data, respectively.) The slope of these curves gives the sensitivity to temperature. Since the automatic high temperature indicator will measure transit time to $\pm$ 0.1μsec, the curves can be used to determine the sensor length required for $\pm$ 50°R resolution (note that 1 in. sensor yields 2 in. path).

The transit times shown are for extensional waves. These times would approximately double for shear waves, i.e., sensor length could be halved, for a given sensitivity.

III. REVIEW OF SHEATH MATERIALS

A. Need for Sheath

A sheath is required to prevent or postpone the undesirable effects of carbon on the sensor. The above test results on 0.030 inch diameter Re wire indicate that this size sensor can be successful in a high temperature graphite environment to the degree that a sheath or protection system prevents carbon diffusion. Without a sheath, this size Re would be usable only up to about 3500°R. With composite W-Re and W sheaths, protection to 4500°R appears attainable, and with a high purity ZrO_2 sheath, protection to 4800°R appears reasonable (see below, Part B.) Sheaths for 0.030 inch diameter Re are discussed next.

B. Sheath Materials for Pure Rhenium

Sheath materials of interest include metals, ceramics, and combinations of the two. Contrary to thermocouple requirements, ceramic sheath (or bead) materials need not be electrically insulating. Two important requirements are chemical stability and carbon barrier action at ~5000°R, preferably up to 1 hour.

1. Metals: W, W-Re, Thoriated W

Tungsten materials presently offer the most reliable carbon diffusion barrier, at least up to 4500°R.* One construction uses a tube of 0.010 inch wall thickness, consisting of a W-25% Re 0.008 inch inner tube surrounded by a 0.002 pure W outer layer. This tube is usable to 4500°R, i.e., to 200°R below its W/Re/C eutectic of 4700°R.

Thoriated tungsten may offer an improved carbon diffusion barrier, but again the eutectic point may limit performance to ~4500°R.

2. Ceramics: BeO, ZrO_2

There is considerable variation in estimates on the maximum usable temperature for ceramics, depending on the application, ceramic purity, time at temperature, etc. Brooks[10] has

*C. D. Pears, Southern Research Institute, private communication (Sept. 26, 1966); G. Remley, WANL, private communication (Sept. 26, 1966); B. Goodier, Los Alamos, private communication (Sept. 26, 1966); W. E. Maurer, Wah Chang, private communication (August 1966).

recently reported that thoria offers the best chemical stability near 5000°R. Hall and Spooner,[11] and others,[12, 13] however, recommend BeO. Union Carbide's experiments,[12] on the contrary, indicate ZrO_2 is usable to higher temperature in contact with carbon, than any other oxide they tested, as shown in the following table.

Refractory Oxide	Practical Limit of Stability for 1 Hour	
$ZrSiO_4$	1675°C	3500°R
CeO_2	1800	3700
Y_2O_3	1900	3900
Al_2O_3	1950	4000
BeO	2100	4270
$SrZrO_3$	2100	4270
ThO_2	2100	4270
HfO_2	2300	4630
ZrO_2	2400	4800

(Source: Union Carbide, DLS-26 (Aug. 1964); ML-TDR-64-125, Vol. II)

We have tested a ZrO_2 closed-end thermowell-type sheath, 1/4 inch O. D. x 5/32 inch I. D. x 8 inches long, in which ZrO_2 powder and a 0.040 inch diameter Mo sensor were packed, to 4200°R in our graphite oven. A purer form of ZrO_2 might survive to higher temperatures, according to the Union Carbide results.

C. Use of Large Diameter Sensor Instead of Sheath

Instead of sheathing, an alternative approach to minimize carbon effects is to use a large diameter sensor. To investigate this approach, ~0.1 inch diameter wires of Mo, Re and 2% thoriated tungsten were obtained and tested ultrasonically at room temperature. Test frequencies from 0.5 to 10 MHz were used, from which 0.5 MHz was found to be the optimum. Lower frequencies could also be used, especially for sensor lengths of ~5 inches, but tests below 0.5 MHz would require the transducer to be modified, and would also require impedance matching transitions between transducer and sensor. The tests at 0.5 MHz used a Branson type ZR piezoelectric transducer which yields a ringing rf pulse at least 30 μsec long, not the 1 to 3 μsec video pulse required for use with the automatic instrument. Accordingly, further work on large diameter sensors was temporarily

postponed, in favor of a concentrated effort on the small diameter sensor, ~0.030 inch diameter.

In conclusion, it appears that additional high temperature ultrasonic testing is required to determine the optimum sheath combination for this ultrasonic application.

IV. AUTOMATIC THIN WIRE ULTRASONIC THERMOMETER

The automatic thin wire ultrasonic thermometer (Figure 9) has now been tested at elevated temperature with satisfactory operation. In preliminary tests, tungsten was heated in air to ~4300°R with an oxy-acetylene torch, with no loss of signal. Similarly, Mo was heated to ~4000°R with no attenuation. These temperatures were obtained from the measured transit time (digital display, to $\pm$ 0.1 μsec) and the known velocity-temperature curve for these wires. In the absence of attenuation, the automatic instrument appears suitable for use with metal wire sensors up to their melting points. Thermometry up to this limit has been shown feasible by Bell et al. in recent work on Cb up to ~5000°R,[13] as well as in Bell's unpublished work on Mo up to its melting point, ~5200°R.

The detailed operation of the automatic ultrasonic thermometer is given in a separate manual. Here it suffices to note that (1) the new unit uses single pulses (not double pulses, as in Bell's manually operated instrument); (2) transit time, averaged over ten measurements, is displayed in digital and analog form, accurate to $\pm$ 0.1 μsec; (3) transit time in the sensor may range up to 999.9 μsec (20 to 30 μsec is more typical, however, for ~2 inch sensors); (4) lead-in length may be 1 to 15 feet; and (5) pulse widths of 1, 2, 3 μsec or, using an external capacitor, > 1 or $\ll$ μsec, are available.

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12. Union Carbide Corp., DLS-26, Improved Graphite Materials for High Temperature Aerospace Use, Vol. II, Development of Graphite Refractory Composites (August 1964); MIL-TDR-64-125, Vol. II, under Contract AF 33(657)-11171.

13. J. F. W. Bell, B. P. Doyle and B. S. Smith, An instrument for the measurement of acoustic pulse velocity and attenuation in a solid probe, J. Sci. Instrum. 43 (1) 28-31 (1966).

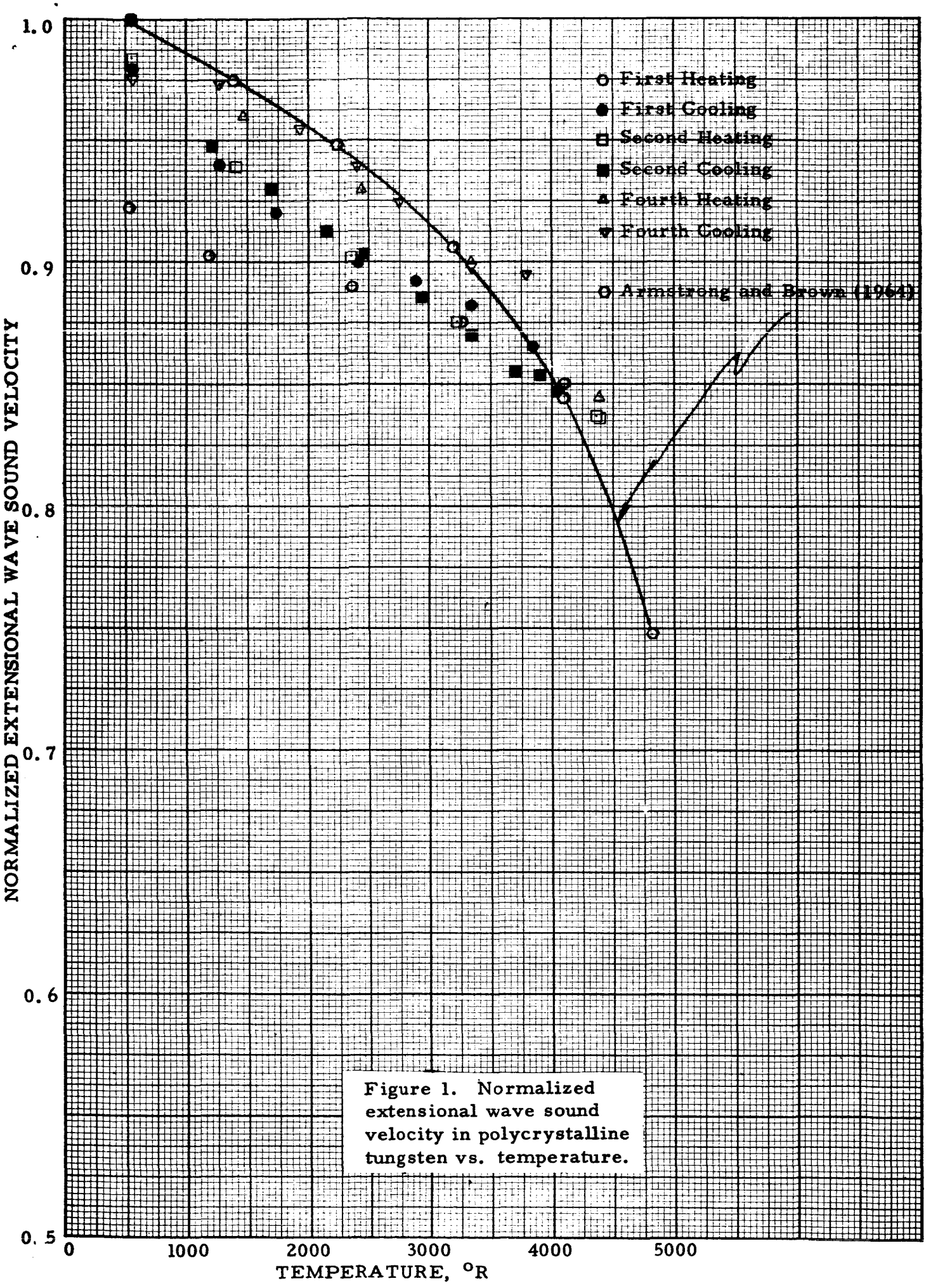
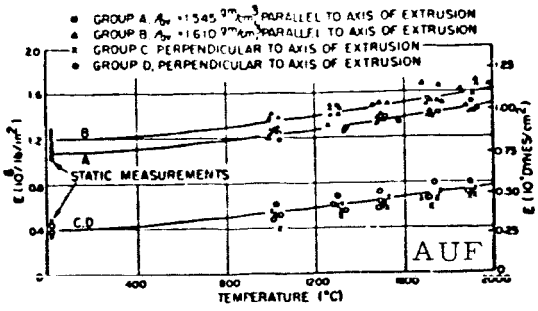
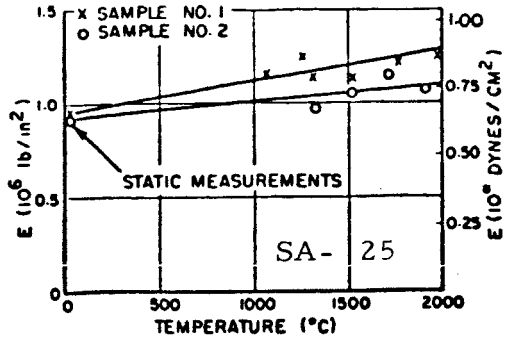


Figure 1. Normalized extensional wave sound velocity in polycrystalline tungsten vs. temperature.



(a) Temperature variation of Young's modulus for AUF graphite.



(b) Temperature variation of Young's modulus for SA-25 graphite.

Fig. 2. Modulus vs. temperature in AUF and SA-25 graphites, after Faris, Green and Smith (1952).

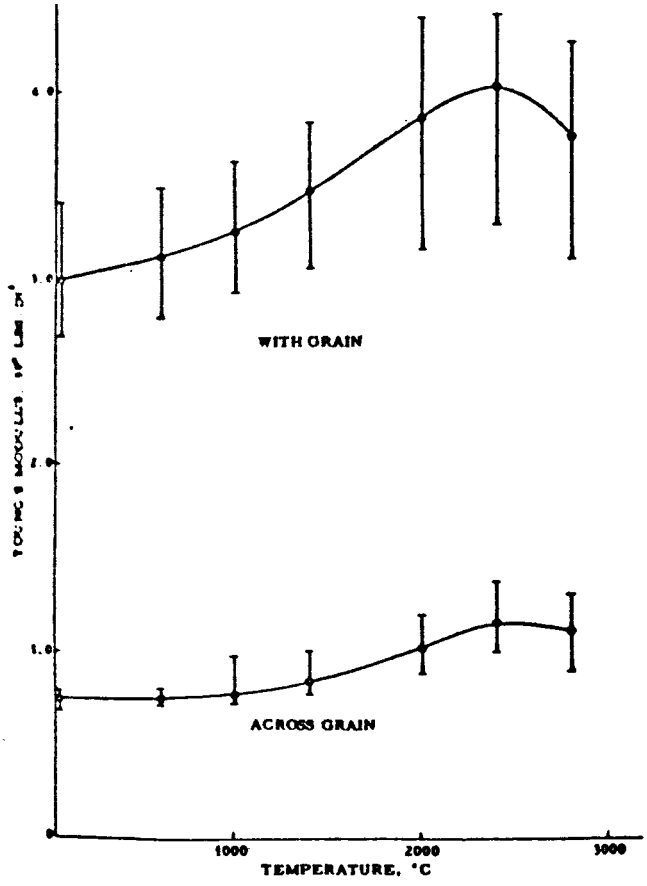


Fig. 3. Young's Modulus Versus Temperature of a Highly Anisotropic Graphite, after Johnson and Dull (1963).

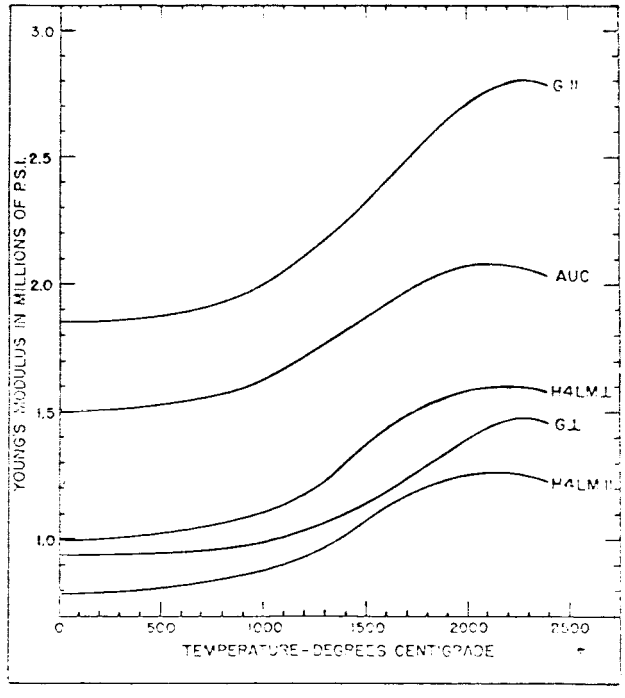


Fig. 4. Young's modulus vs temperature for representative commercial graphites, after Armstrong and Brown (1964).

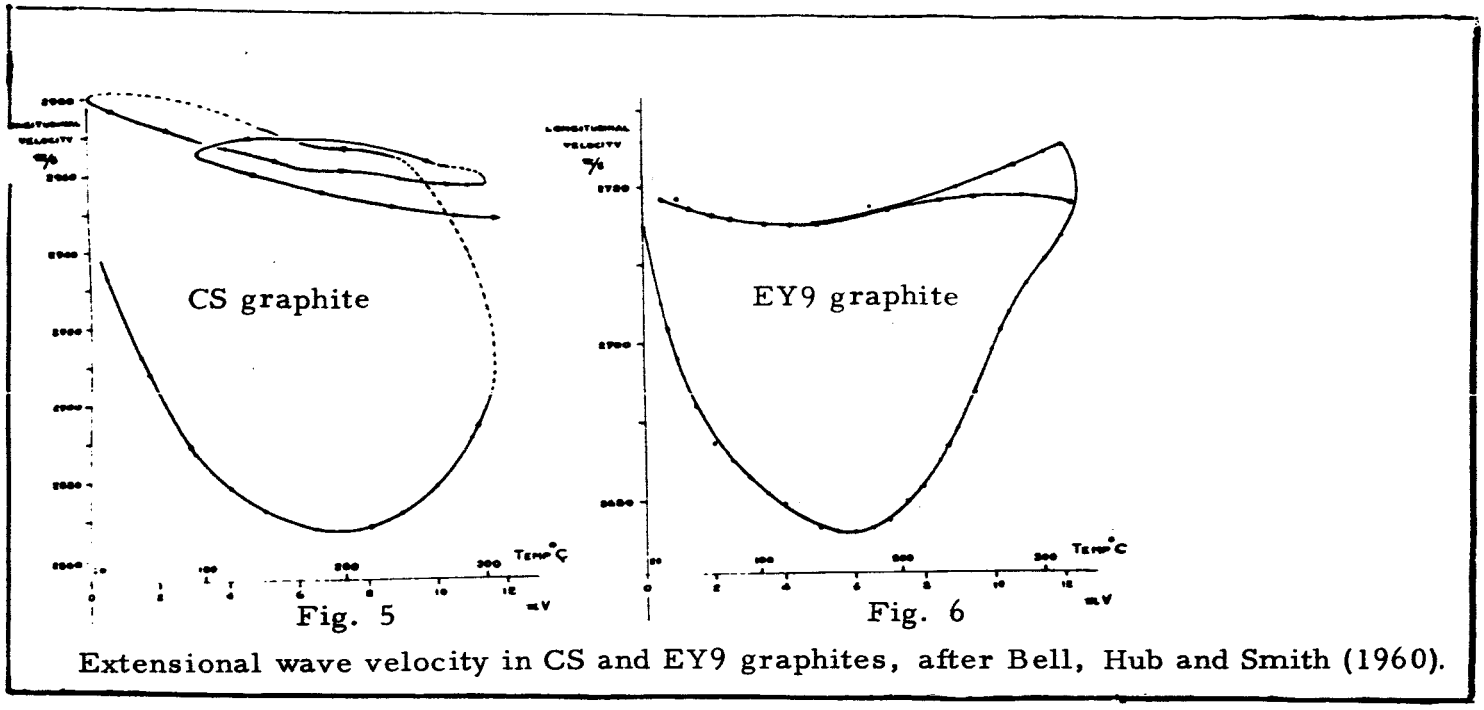


Fig. 5 Fig. 6

Extensional wave velocity in CS and EY9 graphites, after Bell, Hub and Smith (1960).

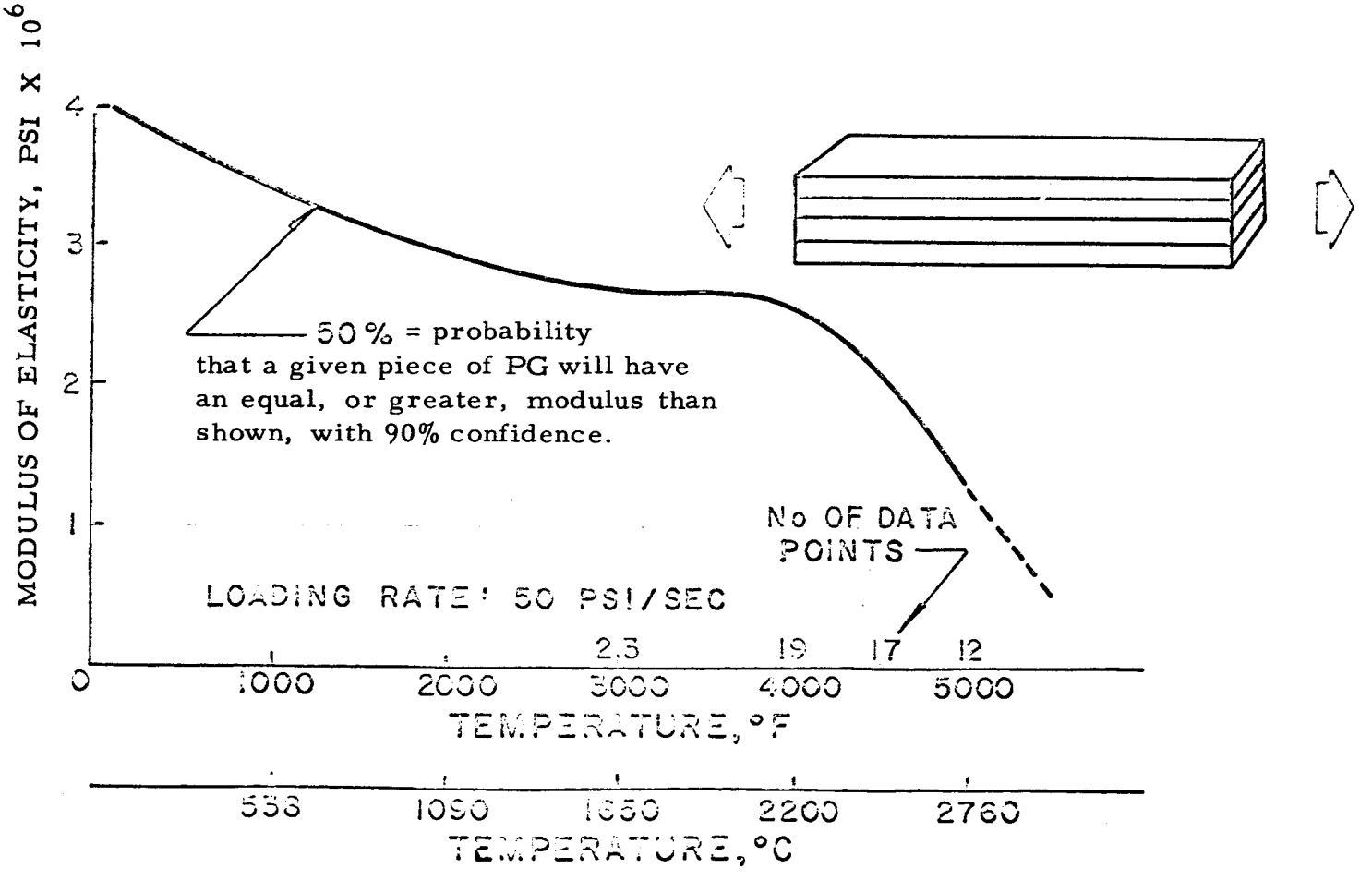


Fig. 7. Modulus of elasticity in tension vs. temperature, in pyrolytic graphite, "a" direction, after Lockheed Missiles and Space Company, LMSC 288186 (1961).

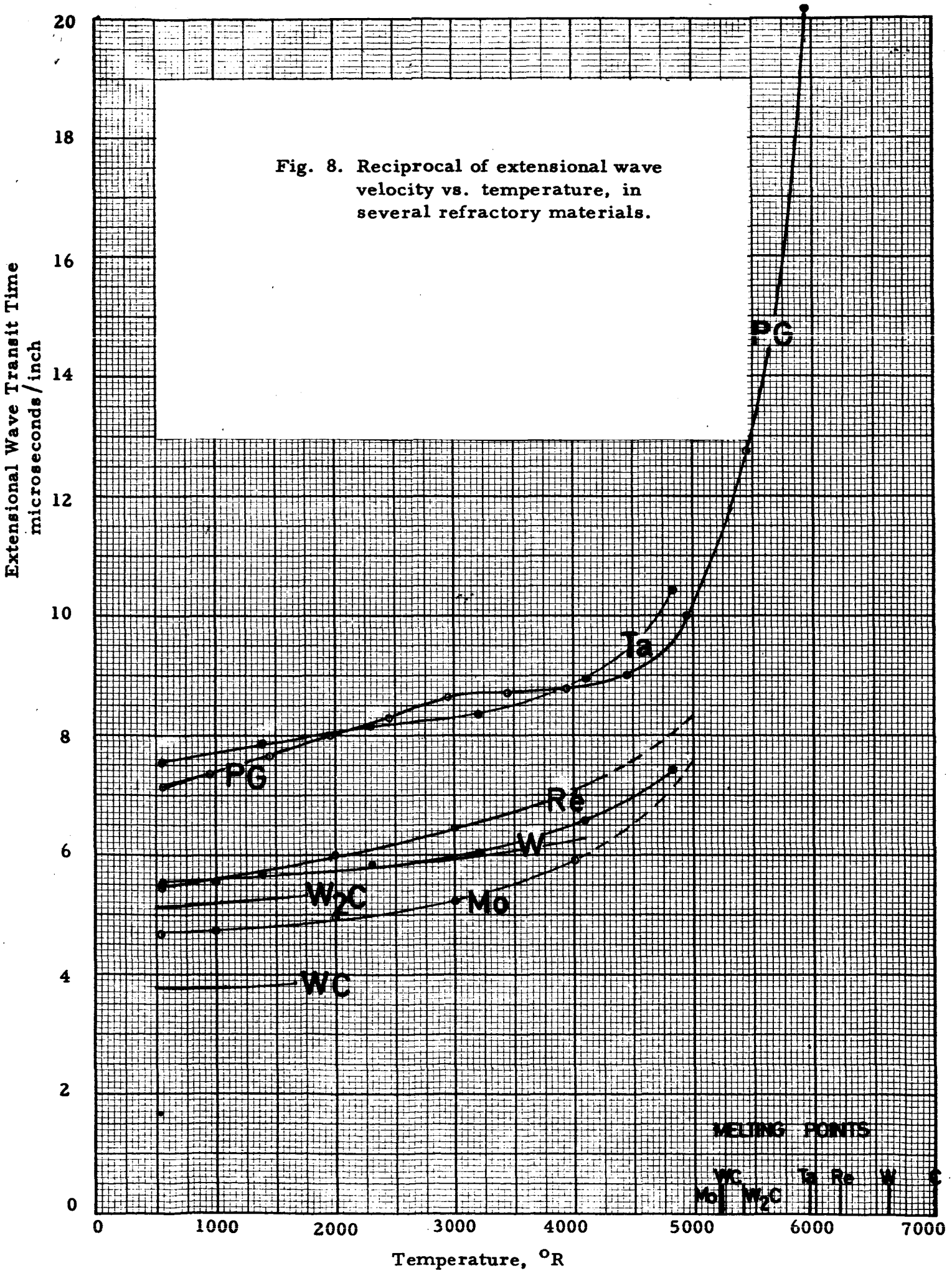


Fig. 8. Reciprocal of extensional wave velocity vs. temperature, in several refractory materials.

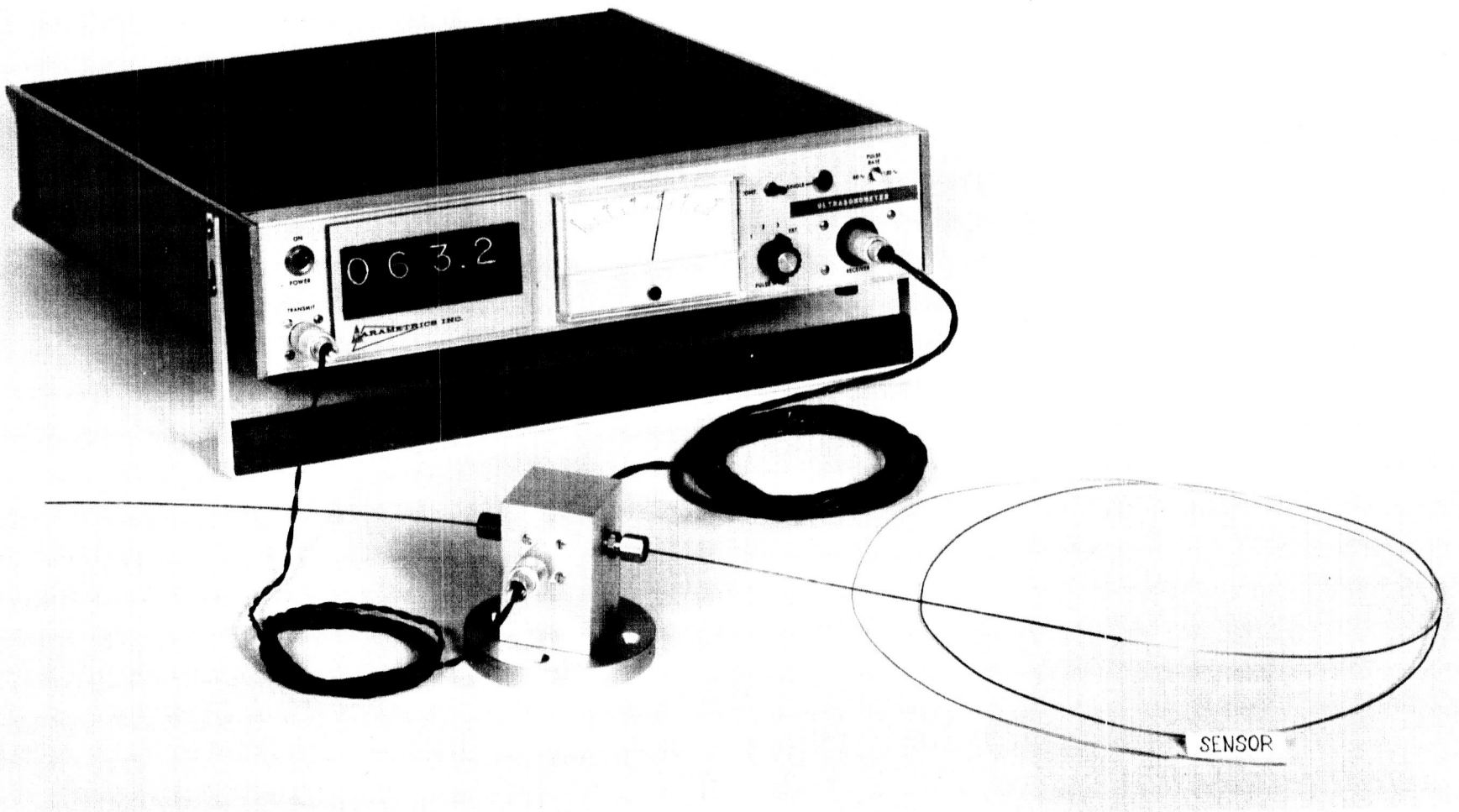


Figure 9. Ultrasonometer automatically measures transit time in sensor, from which temperature is determined. The ultrasonic temperature measuring system illustrated here consists of the Ultrasonometer transmitter/receiver and readout instrument, transducer, lead-in wire and sensor.